Anonymous Referee #3

We greatly appreciate the reviewer's perceptive and constructive comments. Not only did they help to clarify the presentation of our work but also prevent the use of erroneous data in analysis. We have addressed all of the comments carefully, as detailed below.

This is a very interesting manuscript that demonstrates the significant impact of large-scale circulation on trace gases in an urban area, especially GEM. This innovative work yields surprising results that show the importance of meteorology and how it can dominate over anthropogenic emission sources; a surprising result to some. Except for a few minor comments, the manuscript is ready for peer review.

p. 6, line 111 – what type of catalytic converter was used on the TEI42C? If it was the usual moly (molybdenum) converter from TEI, it actually measures NOy not NO2. The Moly converter efficiently (100%) converts NOy species to NO. A blue light converter will provide much more accurate measurements of NO_2

The TEI42C uses a moly converter. The converter is located in the analyzer, not mounted at 10 m high up on a tower, so technically it does not measure NO_y. The moly convertor measures NO_2 plus some fraction of NOz (=HNO_3+HONO+N_2O_5+organic nitrates+…). Dr. Jim Schwab of SUNY at Albany recently gave a presentation (http://www.nescaum.org/documents/nyc-metro-area-energy-air-quality-data-gaps-workshop) that suggests that in NYC (Queens College), that NO_z can be on the order of 12% of the total NO_y, which means that NO+NO_2 is about 88% on average. The reviewer is correct that the method is not specific to NO_2. This is a shortcoming of the method. For the purposes of NAAQS compliance in urban areas, it is not of a major concern to NYS DEC who is responsible for all the monitoring work. However, for the purposes of detailed NO_y budget analyses, it can make a difference. In this study, we used NO_2 as a tracer for fuel combustion sources, so the NO_2 data, albeit imperfect, served the purpose to a large extent. This information was added in the data and approach section for clarification. See lines 121-126

p. 9, lines 192 & 193 - these are surprisingly rapid increases in GEM.

p. 10, line 201 – this is an impressive increase in GEM. Was the calibration checked to ensure no issues with it that might have caused this change? I have never seen anything like this before. The reproducible sinusoidal fluctuations over several hours look to me like an instrument problem. What else could explain these? They are very pronounced and have characteristics of temperature fluctuations with where the instrument was housed. I would double-check these things.

After several discussions with the New York State Department of Conservation (NYSDEC), who operates the site, and the Atmospheric Mercury Network (AMNet), who QA&QC the data and was not aware of the problem previously, it was recommended that the summer 2014 data be removed because the 3-hour sinusoidal fluctuations appeared to be related to a temperature artifact. However, the removal of the data did not change the fundamental findings of the study, as GEM levels at the site were increasing after the lowest point in winter 2011 through spring

2015 (Fig. 3). Most importantly the increases in the seasons from winter 2014 through spring 2015 were consistent with the dynamical analysis.

p. 10, line 213 – what type of meteorological circulations would have caused this increase, and where did such a large source of GEM originate?

In line 213, we hypothesized that the striking contrast of GEM levels in Bronx between winter 2010 and winter 2014 was predominantly caused by the interannual variation in large scale circulation over the region with a particularly strong and persistent influence of the subtropical high pressure system in 2014 – 2015. The northeastern U.S. is a region with relatively concentrated emissions of Hg among other pollutants. In general, one key factor for the Northeastern U.S. to stay relatively clean is strong ventilation, which is facilitated by dynamic systems such as cold frontal passages causing high wind often accompanied by precipitation. Meteorologically this region is under the influence of the Eastern U.S. coastal trough on the 500 hPa pressure level. The large variation in the position and intensity of this trough and naturally the subtropical high pressure system resulted in close association between frontal passages and the subtropical high. A frontal passage wipes polluted air off the continent and bring in relatively clean, whereas a strong influence of the subtropical high produces relatively stagnant conditions conducive to regional build-up of pollution. Our analysis suggested a persistent and strong influence of the subtropical high (i.e. Bermuda High) starting in winter 2014, lasting through the whole year of 2014 (now with summer removed), and extending to spring 2015, as supported by what was shown in Figures 5 – 8. Surface wind data showed in winter 2014 the least frequent and slowest northwesterly wind that could bring relatively clean Canadian air to the region and in summer 2014 the highest frequency of $<1 m\ s^{-1}$ wind (Fig. 5) indicating fairly calm conditions prevailing in the region during the entire season. Further examination of large scale circulation patterns suggested the East U.S. positioned near the axis to the front of the trough in winter 2014, backed by the most negative TAI and TII indices (Fig. 6d) as well as 500 hPa geopotential height and sea-level pressure maps (Figs. 6c,g).

p. 19 – don't these significant decreases in SO_2 and NO_2 emissions additionally rule out these same sources as being important for GEM?

We agree with the reviewer that significant decreases in SO_2 and NO_2 emissions would have indicated the diminishing importance of the common sources of SO_2, NO_2, and GEM. However, the US EPA emission inventories showed emissions of Hg changed in different sectors and in the opposite direction compared to changes in emissions of SO_2 and NO_2 from 2008 to 2011 and from 2011 to 2014. Specifically, NYC Hg anthropogenic emissions were increased by 16% from 2008 to 2011, mainly in miscellaneous non-industrial NEC and waste disposal emissions, and further increased by 37% from 2011 to 2014 primarily in fuel combustion. Eastern U.S. emissions of Hg decreased by 13% from 2008 to 2011 and increased by 2% from 2011 to 2014. In contrast, NYC SO_2 emissions decreased steadily by 30% from 2008 to 2011 followed by a further decrease of 43% to 2014, while Eastern U.S. emissions decreased by 48% from 2008 to 2011 and furthered by another 29% decrease in 2014. As for NO_2, fuel and mobile combustion emissions comprised >99.5% of the total NO_x emissions in NYC and ~90% over the Eastern US. NYC NO_x emissions changed insignificantly (1%) from 2008 to 2011 and decreased by 15% from 2011 to 2014, while Eastern US mobile and fuel combustion emissions were decreased by

16% and 33%, respectively, from 2008 to 2011, and further decreased by 13% and 9%, respectively, to 2014. Therefore, changes in SO_2 and NO_2 emissions could not necessarily indicate the direction or magnitude of changes in Hg emissions.